

Attorney's Docket: 1999DE132  
Serial No.: 09/722,760  
Group: 1758

### AMENDMENTS TO THE CLAIMS

1. (Currently Amended) A method of imparting, controlling or improving the charge of an electrophotographic toner or developer, or an electret material, comprising the ~~step of adding as a charge control agent a structured silicate salt which contains~~ wherein the cation is a low molecular weight organic cation or a combination of a low molecular weight organic cation with  $\text{NH}_4^+$ ,  $\text{H}_3\text{O}^+$ , an alkali metal, an alkaline earth metal, an earth metal or with a transition metal and the anion is an island, cyclic, group, chain, ribbon, laminar or matrix silicate or a combination thereof to a binder of an electrophotographic toner or developer or of an electret material.

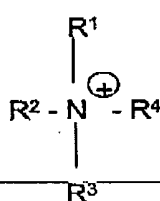
2) (Cancelled)

3) (Cancelled)

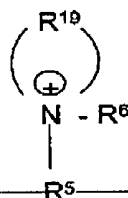
4) (Currently Amended) The method as claimed in claim 1, wherein the low molecular weight organic cation is a substituted ~~ammonium~~, phosphonium, thionium or triphenylcarbonium ion or a cationic metal complex.

5) (Currently Amended) The method as claimed in ~~claim 4~~claim 1, wherein the low molecular weight organic cation is an ammonium ion ~~has having~~ one of the formulae (a) - (j)

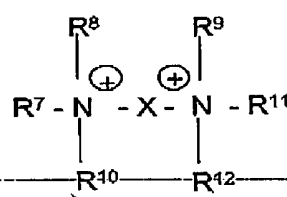
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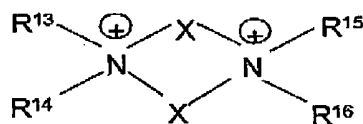
(a)



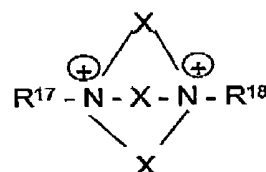
(b)



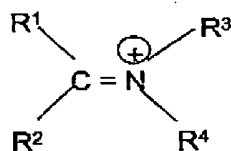
(c)



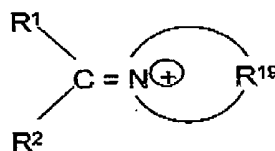
(d)



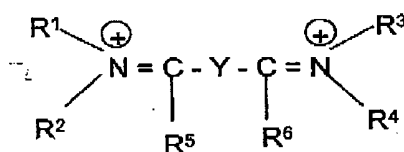
(e)



(f)

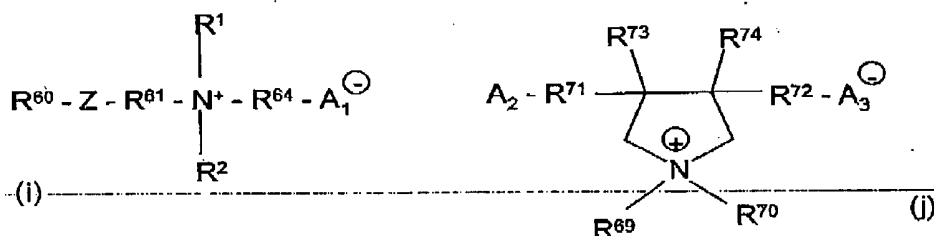


(g)



(h)

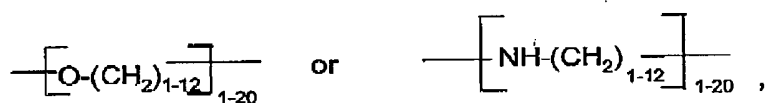
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in which

$\text{R}^1$  to  $\text{R}^{18}$  are identical or different and represent hydrogen, CN,  $(\text{CH}_2)_{1-16}\text{CN}$ , halogen, branched or unbranched  $\text{C}_1$ - $\text{C}_{32}$ -alkyl, mono- or polyunsaturated  $\text{C}_2$ - $\text{C}_{32}$ -alkenyl,  $\text{C}_1$ - $\text{C}_{22}$ -alkoxy,  $\text{C}_1$ - $\text{C}_{22}$ -hydroxyalkyl,  $\text{C}_1$ - $\text{C}_{22}$ -halogenoalkyl,  $\text{C}_2$ - $\text{C}_{22}$ -halogenoalkenyl,  $\text{C}_1$ - $\text{C}_{22}$ -aminoalkyl,  $(\text{C}_1$ - $\text{C}_{12}$ )-trialkyl-ammonium- $(\text{C}_1$ - $\text{C}_{22}$ )-alkyl;  $(\text{C}_1$ - $\text{C}_{22}$ )-alkylene- $(\text{C}=\text{O})\text{O}$ - $(\text{C}_1$ - $\text{C}_{32}$ )-alkyl,  $(\text{C}_1$ - $\text{C}_{22}$ )-alkylene- $(\text{C}=\text{O})\text{O}$ -aryl,  $(\text{C}_1$ - $\text{C}_{22}$ )-alkylene- $(\text{C}=\text{O})\text{NH}$ - $(\text{C}_1$ - $\text{C}_{32}$ )-alkyl,  $(\text{C}_1$ - $\text{C}_{22}$ )-alkylene- $(\text{C}=\text{O})\text{NH}$ -aryl,  $(\text{C}_1$ - $\text{C}_{22}$ )-alkylene- $\text{O}(\text{CO})$ - $(\text{C}_1$ - $\text{C}_{32}$ )-alkyl,  $(\text{C}_1$ - $\text{C}_{22}$ )-alkylene- $\text{O}(\text{CO})$ -aryl,  $(\text{C}_1$ - $\text{C}_{22}$ )-alkylene- $\text{NH}(\text{C}=\text{O})$ - $(\text{C}_1$ - $\text{C}_{32}$ )-alkyl,  $(\text{C}_1$ - $\text{C}_{22}$ )-alkylene- $\text{NHCO}$ -aryl,

wherein



are optionally inserted into the acid ester or acid amide bonds;

$[(\text{C}_1$ - $\text{C}_{12})$ -alkylene- $\text{O}]_{1-100}$ -H; aryl,  $(\text{C}_1$ - $\text{C}_{18})$ -alkylenearyl;  $-(\text{O}-\text{SiR}'_2)_{1-32}-\text{O}-\text{SiR}'_3$ , in which  $\text{R}'$  has the meaning  $\text{C}_1$ - $\text{C}_{12}$ -alkyl, phenyl, benzyl or  $\text{C}_1$ - $\text{C}_{12}$ -alkoxy; heterocyclyl,  $\text{C}_1$ - $\text{C}_{18}$ -alkylene-heterocyclyl, wherein the aryl and heterocyclyl radicals are optionally mono- or polysubstituted on carbon atoms or heteroatoms by  $\text{C}_1$ - $\text{C}_{12}$ -alkyl,  $\text{C}_1$ - $\text{C}_4$ -alkenyl,  $\text{C}_1$ - $\text{C}_4$ -alkoxy, hydroxy- $(\text{C}_1$ - $\text{C}_4$ )-alkyl, amino- $(\text{C}_1$ - $\text{C}_4$ )-alkyl,  $\text{C}_1$ - $\text{C}_4$ -alkylimino, carboxyl, hydroxyl, amino, nitro, cyano, halogen,  $\text{C}_1$ - $\text{C}_{12}$ -acyl,  $\text{C}_1$ - $\text{C}_4$ -halogenoalkyl,  $\text{C}_1$ - $\text{C}_4$ -alkylcarbonyl,  $\text{C}_1$ - $\text{C}_4$ -alkylcarbonyloxy,  $\text{C}_1$ - $\text{C}_4$ -alkoxycarbonyl,  $\text{C}_1$ - $\text{C}_4$ -alkylaminocarbonyl,  $\text{C}_1$ - $\text{C}_4$ -alkylcarbonylimino,  $\text{C}_6$ - $\text{C}_{10}$ -arylcarbonyl, aminocarbonyl, aminosulfonyl,  $\text{C}_1$ - $\text{C}_4$ -alkylaminosulfonyl, phenyl, naphthyl, or heteroaryl;

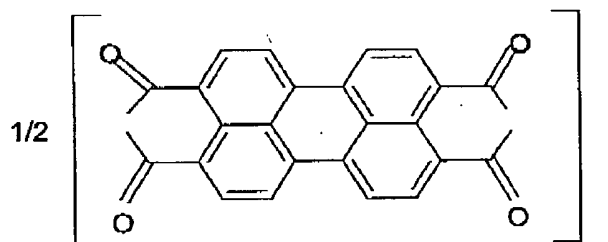
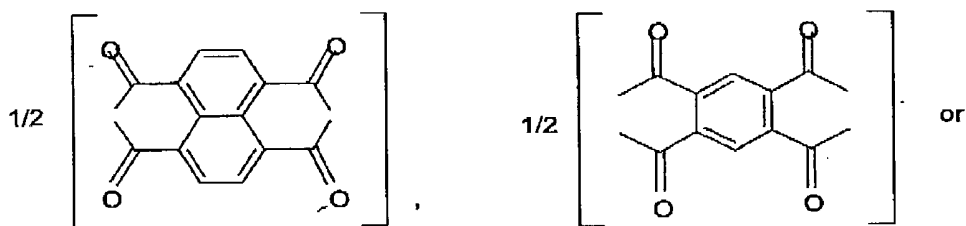
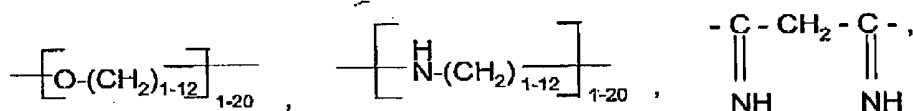
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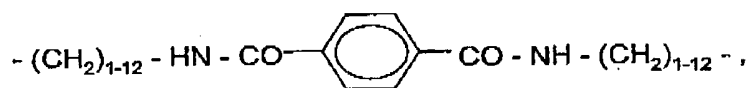
$R^{19}$  represents  $C_4$ - $C_{11}$ -alkylene,  $-(C_2H_4-O-)_{1-17}(CH_2)_{1-2-}$ ,  $-(C_2H_4-NR-)_{1-17}(CH_2)_{1-2-}$ , in which R is hydrogen or  $C_1$ - $C_{12}$ -alkyl;

X has the meaning of Y or  $-CO-CH_2-CO-$ ,



Y has the meaning  $\begin{array}{c} -C- \\ || \\ O \end{array}$ ,  $\begin{array}{c} -C- \\ || \\ S \end{array}$ ,  $\begin{array}{c} -C- \\ || \\ NH \end{array}$ ,  $-(CH_2)_{1-18}-$ ,

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or o-, p-, m-(C<sub>6</sub>-C<sub>14</sub>)-arylene or (C<sub>4</sub>-C<sub>14</sub>)-heteroarylene with 1, 2, 3 or 4 heteroatoms selected from the group consisting of N, O, S and a combination thereof;

R<sup>60</sup> represents C<sub>1</sub>-C<sub>32</sub>-acyl, C<sub>1</sub>-C<sub>22</sub>-alkyl, C<sub>2</sub>-C<sub>22</sub>-alkenyl, C<sub>1</sub>-C<sub>18</sub>-alkylene-C<sub>6</sub>-C<sub>10</sub>-aryl, C<sub>1</sub>-C<sub>22</sub>-alkylene-heterocyclyl, C<sub>6</sub>-C<sub>10</sub>-aryl or (C<sub>4</sub>-C<sub>14</sub>)-heteroaryl with 1, 2, 3 or 4 heteroatoms selected from the group consisting of N, O, S, and a combination thereof;

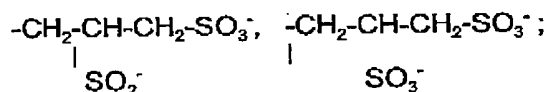
R<sup>61</sup> and R<sup>64</sup> represent -(CH<sub>2</sub>)<sub>1-18</sub>-, C<sub>1</sub>-C<sub>12</sub>-alkylene-C<sub>6</sub>-C<sub>10</sub>-arylene, C<sub>6</sub>-C<sub>10</sub>-arylene, C<sub>0</sub>-C<sub>12</sub>-alkylene-heterocyclyl;

Z represents -NH- or -O-;

A<sub>1</sub> and A<sub>3</sub> represent -COO<sup>-</sup>, -SO<sub>3</sub><sup>-</sup>, -OSO<sub>3</sub><sup>-</sup>, -SO<sub>2</sub><sup>-</sup>, -COS<sup>-</sup> or -CS<sub>2</sub><sup>-</sup>;

A<sub>2</sub> represents -SO<sub>2</sub>Na, -SO<sub>3</sub>Na, -SO<sub>2</sub>H, -SO<sub>3</sub>H or hydrogen;

R<sup>68</sup> and R<sup>70</sup> independently of one another represent hydrogen, C<sub>1</sub>-C<sub>32</sub>-alkyl, in which the alkyl chain optionally contain one or more of the groups -NH-CO-, -CO-NH-, -CO-O- or -O-CO-; C<sub>1</sub>-C<sub>18</sub>-alkylene-aryl, C<sub>0</sub>-C<sub>18</sub>-alkylene-heterocyclyl, C<sub>1</sub>-C<sub>18</sub>-hydroxyalkyl, C<sub>1</sub>-C<sub>18</sub>-halogenoalkyl, aryl, -(CH<sub>2</sub>)<sub>3</sub>-SO<sub>3</sub><sup>-</sup>,



R<sup>71</sup> and R<sup>72</sup> represent -(CH<sub>2</sub>)<sub>1-12</sub>-; and

R<sup>73</sup> and R<sup>74</sup> represent hydrogen or C<sub>1</sub>-C<sub>22</sub>-alkyl.

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6) (Original) The method as claimed in claim 5, wherein  $R^1$  to  $R^{18}$  denote hydrogen, CN,  $\text{CH}_2\text{-CN}$ ,  $\text{CF}_3$ ,  $\text{C}_1\text{-C}_{22}\text{-alkyl}$ ,  $\text{C}_2\text{-C}_{18}\text{-alkenyl}$ ,  $\text{C}_1\text{-C}_{18}\text{-alkoxy}$ ,  $\text{C}_1\text{-C}_{18}\text{-hydroxy-alkyl}$ ,  $\text{C}_1\text{-C}_{18}\text{-halogenoalkyl}$ ,  $\text{C}_2\text{-C}_{18}\text{-halogenoalkenyl}$ ,  $\text{C}_1\text{-C}_{18}\text{-aminoalkyl}$ ,  $(\text{C}_1\text{-C}_6)\text{-trialkylammonium-(C}_1\text{-C}_{18}\text{-alkyl)}$ ,  $(\text{C}_1\text{-C}_{18}\text{-alkylene-O(C=O)-(C}_1\text{-C}_{22}\text{-alkyl)}$ ,  $(\text{C}_1\text{-C}_{18}\text{-alkylene-O(C=O)-phenyl}$ ,  $(\text{C}_1\text{-C}_{18}\text{-alkylene-NHCO-(C}_1\text{-C}_{22}\text{-alkyl)}$ ,  $(\text{C}_1\text{-C}_{18}\text{-alkylene-NHCO-phenyl}$ ,  $(\text{C}_1\text{-C}_{18}\text{-alkylene-(C=O)O-(C}_1\text{-C}_{22}\text{-alkyl)}$ ,  $(\text{C}_1\text{-C}_{18}\text{-alkylene-(C=O)O-phenyl}$ ,  $(\text{C}_1\text{-C}_{18}\text{-alkylene-(C=O)NH-(C}_1\text{-C}_{22}\text{-alkyl)}$ ,  $(\text{C}_1\text{-C}_{18}\text{-alkylene-CONH-phenyl}$ , benzyl, phenyl, naphthyl,  $\text{C}_1\text{-C}_{12}\text{-alkylene-heterocyclyl}$ ;  
 $R^{19}$  denotes  $\text{C}_4\text{-C}_5\text{-alkylene}$ ,  $-(\text{C}_2\text{H}_4\text{-O})_{1-9}\text{-(CH}_2\text{)}_{1-2}$  or  $-(\text{C}_2\text{H}_4\text{-NH})_{1-9}\text{-(CH}_2\text{)}_{1-2}$ ;  
 $R^{60}$  denotes  $\text{C}_1\text{-C}_{18}\text{-acyl}$ ,  $\text{C}_1\text{-C}_{18}\text{-alkyl}$ ,  $\text{C}_2\text{-C}_{18}\text{-alkenyl}$ ,  $\text{C}_1\text{-C}_{12}\text{-alkylene-phenyl}$ ,  $\text{C}_1\text{-C}_{18}\text{-alkylene-pyridyl}$ , phenyl or pyridyl;  
 $R^{61}$  and  $R^{64}$  denote  $-(\text{CH}_2)_{1-12}$ ,  $\text{C}_1\text{-C}_8\text{-alkylene-phenylene}$ , phenylene or  $\text{C}_1\text{-C}_8\text{-alkylenepyridylene}$  or piperidylene;  
 $R^{71}$  and  $R^{72}$  denote  $-(\text{CH}_2)_{1-8}$  and  
 $R^{73}$  and  $R^{74}$  denote hydrogen or  $(\text{C}_1\text{-C}_{18})\text{-alkyl}$ .

7) (Currently Amended) The method as claimed in ~~claim 4~~ claim 1, wherein the low molecular weight organic cation is an ammonium ion which is an aliphatic or aromatic 5- to 12-membered heterocyclic radical with 1 to 4 atoms selected from the group consisting of N, O and S, or a combination thereof, belonging to the rings.

8) (Original) The method as claimed in claim 7, wherein the heterocyclic radical is pyridinium, pyridazinium, pyrimidinium, pyrazinium, purinium, tetraazaporphyrinium, piperidinium, morpholinium, tetrazonium, triaza-cyclononanium or tetraaza-cyclododecanium.

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9) (Original) The method as claimed in claim 4, wherein the cationic metal complex is a metal carboxylate, metal salicylate, metal sulfonate, 1:1 metal-azo complex or a metal dithiocarbamate.

10) (Previously Presented) The method as claimed in claim 9, wherein the metal is selected from the group consisting of Al, Mg, Ca, Sr, Ba, TiO, VO, Cr, V, Ti, Zr, Sc, Mn, Fe, Co, Ni, Cu, Zn and ZrO.

11) through 15) (Cancelled)

16) (Previously Presented) The method of claim 4, wherein the ammonium ion is an aliphatic or aromatic 5- to 12-membered heterocyclic radical with 1 to 4 atoms selected from the group consisting of N, O and S, or a combination thereof, belonging to the rings, wherein 2 to 8 rings are fused.

17) (Previously Presented) The method as claimed in claim 9, wherein the metal is selected from the group consisting of Al, Mg, Ca, Sr, Ba, TiO, VO, Cr, V, Ti, Zr, Sc, Mn, Fe, Co, Ni, Cu, Zn and ZrO, and the metal complex contains one or more further ligands.

18) through 21) (Cancelled)

22) (Currently Amended) A method of imparting, controlling or improving the charge of an electrophotographic toner or developer, or an electret material comprising the step of adding a distearyldimethyl ammonium bentonite charge control agent to a binder of an electrophotographic toner or developer or of an electret material, wherein the charge control agent is distearyldimethyl ammonium bentonite.

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23) (New) A method of imparting, controlling or improving the charge of an electrophotographic toner or developer, or an electret material, comprising the step of adding as a charge control agent a structured silicate salt wherein the cation is a low molecular weight organic cation or a combination of a low molecular weight organic cation with  $\text{NH}_4^+$ ,  $\text{H}_3\text{O}^+$ , an alkali metal, an alkaline earth metal, an earth metal or with a transition metal and the anion is an island, cyclic, group, chain, ribbon, laminar or matrix silicate or a combination thereof to a binder of an electrophotographic toner or developer or of an electret material, wherein the charge control agent imparts either a positive or negative charge.